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"Cut-and-paste" method for the rapid prototyping of soft electronics

YANG XiangXing[1], HUANG YiFu[1], DAI ZhaoHe[2], BARBER Jamie[1], WANG PuLin[3] & LU NanShu[1,2,4,5*]

[1]*Department of Electrical and Computer Engineering, the University of Texas at Austin, Austin 78712, USA;*
[2]*Center for Mechanics of Solids, Structures and Materials, Department of Aerospace Engineering and Engineering Mechanics, the University of Texas at Austin, Austin 78712, USA;*
[3]*Stretch Med Inc., Austin 78750, USA;*
[4]*Department of Biomedical Engineering, the University of Texas at Austin, Austin 78712, USA;*
[5]*Texas Materials Institute, the University of Texas at Austin, Austin 78712, USA*

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Unlike wafer-based rigid electronics, soft electronics have many unique advantages including thinness, flexibility, stretchability, conformability, lightweight, large area, as well as low cost. As a result, they have demonstrated many emerging capabilities in healthcare devices, soft robotics, and human-machine interface. Instead of conventional microfabrication, there is an ever-growing interest in the freeform or digital manufacture of soft electronics. This review provides a survey for a cost- and time-effective subtractive manufacturing process called the "cut-and-paste" method. It employs a mechanical cutter plotter to form patterns on various electronically functional membranes such as sheets of metals, functional polymers, and even two-dimensional (2D) materials, supported by a temporary tape. The patterned membranes can then be pasted on soft substrates such as medical tapes or even human skin. This process is completely dry and desktop. It does not involve any rigid wafers and is hence capable of making large-area electronics. The process can be repeated to integrate multiple materials on a single substrate. Integrated circuits (ICs) and rigid components can be added through a "cut-solder-paste" process. Multilayer devices can also be fabricated through lamination. We therefore advocate that the "cut-and-paste" method is a very versatile approach for the rapid prototyping of soft electronics for various applications.

cut-and-paste, subtractive manufacture, epidermal electronics, electronic tattoos, 2D materials

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1 Introduction

Soft electronics [1], including flexible [2] and stretchable electronics [3], are capable of surviving mechanical deformations such as bending, stretching and twisting, as well as conforming to curvilinear surfaces. Such deformability have enabled next-generation devices beyond wafer-based electronics, which include electronic skins or skin-like electronics [4–10], bio-mimetic electronic eye cameras [11,12], wearable as well as invasive healthcare devices [13–17] and human-machine interfaces [18–21], flexible energy harvesting and storage devices [22–24], flexible displays [25–27], deformable antennas [28–32], and so on. The success of soft electronics is built upon new electronic/optical materials, mechanics-guided novel designs, and unconventional manufacturing processes. The mechanics and materials of soft electronics have been reviewed extensively [1,3,33–37]. Therefore, in this review, we will discuss the

* Corresponding author (email: nanshulu@utexas.edu)

manufacturing approaches for soft electronics.

Inorganic flexible and stretchable electronics were first successfully fabricated by the well-known transfer printing method [38–42]. It is a highly versatile method that leverages conventional lithographic process to form patterns and then transfer these patterned inorganic electronic nanomembranes from rigid handeling wafers to deformable polymer substrates. However, it is well known that lithographic process is expensive and time-consuming, plus the transfer printing process demands exceptional experimental skills. Therefore, it may not be the most suitable process for the rapid prototyping of soft electronics. The recent development of free-form fabrication techniques [43–46] offers new capabilities for the digital manufacturing of soft electronics. Examples include additive strategies (e.g. inkjet printing [47], 3D printing [48,49]) and subtractive strategies (e.g. laser cutting [50,51]). Although additive approaches are under fast development, they are limited by the availability of inks and ink sintering time and temperature. Although laser cutter is capable of high-speed processing plate-like materials, problems associated with thermal stress or even thermal damage become more pronounced when processing ultrathin membranes. An alternative subtractive process named the "cut-and-paste" method employs paper/vinyl cutter plotters to mechanically cut seams on thin membranes or bilayer laminates according to the imported design, followed by the manual removal of the excess regions, and finally pasting the patterns on target soft substrates. No chemical or heat is involved in this process. This method has been demonstrated to be able to pattern various electronically functional membranes such as metal and conductive polymer sheets [28,52,53], ceramic thin films such as indium tin oxide (ITO) [54], and even atomically thin two-dimensional (2D) materials [55,56]. This reiew will focus around the materials, processes, and applications of the "cut-and-paste" method.

This review is organized as follows. Sect. 2 introduces the "cut-and-paste" process for patterning conventional metallic, polymeric, and ceramic thin films. Sect. 3 focuses on using the "cut-and-paste" method to manufacture soft electronics out of emerging 2D materials. Sect. 4 discusses an improved "cut-solder-paste" process to add integrated circuits (IC) and other discrete components to the system. Concluding remarks are offered in Sect. 5.

2 "Cut-and-paste" of thin films

2.1 The concept

The process of using a paper/vinyl cutter plotter to form microstructures was first introduced as "xurography", a.k.a. "razor writing" in 2005 for the rapid prototyping of shadow masks and microfluidic molds [57]. The cutting resolution for different types of thin films including metal sheets, polymeric films and thermal adhesives has been reported [57]. Thereafter, the application of xurography was mainly focused on the fabrication of masks and molds until our group invented the "cut-and-paste" method to build stretchable electronics in 2015 [52]. Stretchability of intrinsically stiff electronic materials can be built into the geometry hence patterning is an indispensable step. We therefore proposed to use cutter plotters to form stretchable patterns in metallic, ceramic, polymeric and 2D materials. This simple yet versatile manufacturing technique was born out of a need for the rapid and low-cost prototyping of soft disposable wearable electronics, in contrast to the expensive and time-consuming photolithography process. The "cut-and-paste" process, in its entirety, is described in the follows. First, rolls of gold and aluminum-coated polyethylene terephthalate (PET) were purchased thus eliminating the need for vacuum metal deposition. Next, the gold-coated PET foil (Au-PET) was attached to a thermal release tape (TRT) with Au facing the adhesive and the TRT attached to a sticky cutting mat as shown in Figure 1(a). The sticky cutting mat was fed into a commercially available mechanical cutter plotter (Silhouette Cameo2), and AutoCAD drawings were imported to program the cuts. Figure 1(b) illustrates a carved pattern. After peeling from the cutting mat (Figure 1(c)), the TRT should be heated up to deactivate the adhesives. For thinner films where strong adhesion is not required during cutting, low adhesive tapes can be used instead of the TRT such that the heating step can be omitted. Tweezers were used to peel away excess Au-PET as shown in Figure 1(d). Finally, the leftover pattern was pasted to the desired substrate, such as a 3M Tegaderm tape, as shown in Figure 1(e). One round of the "cut-and-paste" process can be completed within about 10 min. Multiple rounds can be repeated to integrate multilayers and/or multiple materials onto a single substrate via alignment markers. In addition to polymer-supported metal films, electrically conductive rubber was also cut-and-pasted as a soft wearable strain gauge [52]. The only limitation with the Silhouette Cameo cutter is that the minimum achievable serpentine ribbon width is 200 μm when cutting a bilayer foil of 9-μm-thick Al on 12-μm-thick PET. For different materials of different thicknesses, the blade type, blade setting and cutting speed should be adjusted accordingly to achieve the optimal resolution.

As a demonstration, we fabricated a multi-material, multifunctional epidermal sensor system (ESS) (Figure 1(f) and (g)). It was tested to have an effective modulus of 11 MPa and a stretchability beyond 30%. It could be conformably laminated on any part of human skin to detect a variety of biometrics including electrophysiological signals such as electrocardiogram (ECG), electromyogram (EMG), and electroencephalogram (EEG), skin temperature, skin hydration, and respiratory rate, which have been validated against available commercial tools. The stretchable antenna on the

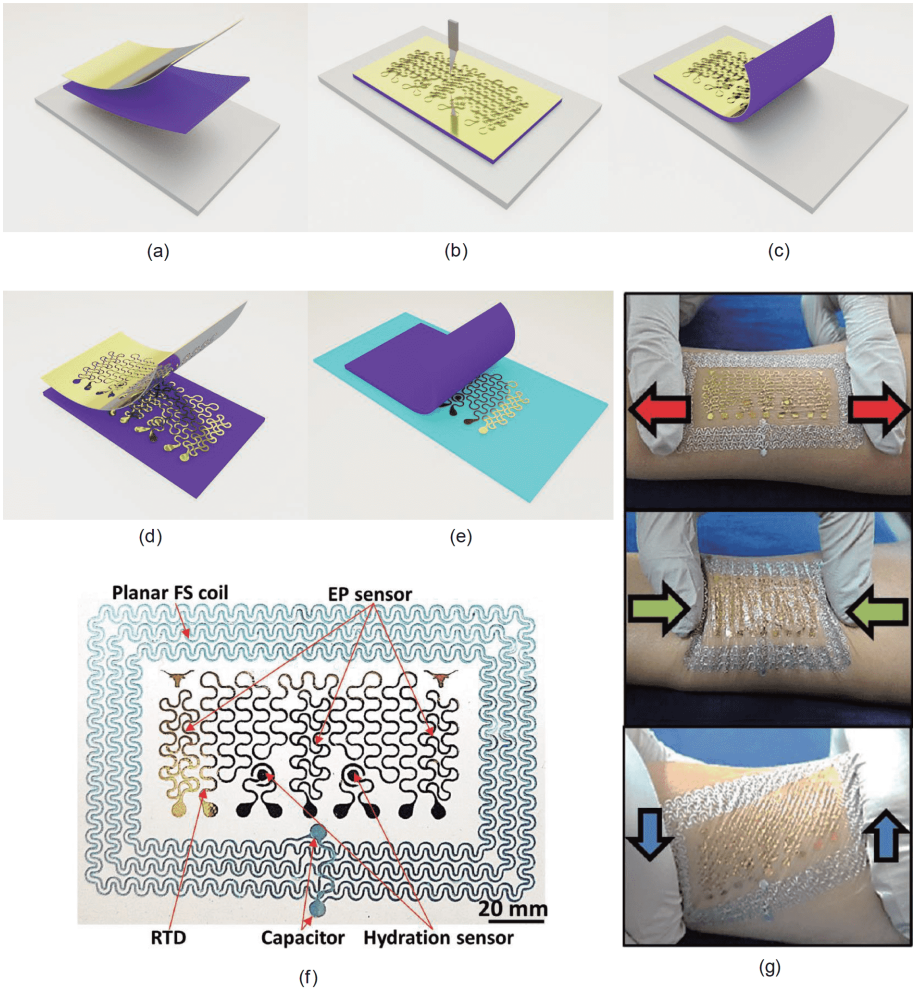

Figure 1 (Color online) Schematics for the "cut-and-paste" process and multi-material, multifunctional epidermal sensor system (ESS). (a) Au-PET-TRT (APT) laminated on the cutting mat; (b) carving designed seams in the Au-PET layer by programmable paper/vinyl cutter plotter; (c) peeling APT off the cutting mat; (d) removing excess Au-PET regions after deactivating the TRT on hot plate; (e) pasting patterned Au-PET onto the target substrate; (f) top view of an ESS which incorporates three electrophysiological (EP) electrodes (Au-PET), a resistance temperature detector (RTD) (Au-PET), two coaxial dot-ring impedance sensors (Au-PET), and a wireless planar stretchable strain sensing coil (Al-PET), all in filamentary serpentine (FS) layout for stretchability; (g) ESS on human skin demonstrating excellent deformability under applied stretch (top), compression (middle), and shear (bottom). Figure 1(f) and (g) is republished with permission from Advanced Materials [52].

ESS could be measured wirelessly and hence could be applied for near field communication (NFC)-enabled wireless data and power transfer in the future. Thus, the "cut-and-paste" method has opened the door for the rapid prototyping of deformable electronics represented by ESS, which is also known as the electronic tattoos (e-tattoos).

In its first development, the "cut-and-paste" method had to use a substrate such as a medical tape to support the stretchable sensors, which increased tattoo thickness and degraded breathability. To address this problem, we came up with a slightly modified "cut-and-paste" method to fabricate tape-free e-tattoos with a total thickness of just 1.5 μm [58]. Instead of using TRT or low adhesive tapes, we employed commercial tattoo papers with water soluble adhesives as the temporary support. After the pattern was formed, the 1.5-μm-thick sensor network can be directly pasted on human skin by applying water on the tattoo paper to dissolve the adhesive. Thanks to its micrometer thinness, the sensor network can adhere to human skin via just van der Waals forces [59,60]. It formed an open-mesh filamentary serpentine network on the skin, which allowed for superior compliance and breathability. Such ultrathin, tape-free e-tattoo has demonstrated clear advantages over previous e-tattoos regarding skin-electrode interface impedance, motion artifacts, and sweat artifacts.

Following our "cut-and-paste" method, Zhou et al. [53] manufactured stretchable and dry surface electromyography (sEMG) electrodes for gesture interpretation. Kao et al. [61] fabricated "Duoskin" out of metal leaves as on-skin user interface. However, without the polymer (e.g. PET) support for the metal leaves, the mechanical robustness of "Duoskin" may be very limited [62,63].

2.2 "Cut-and-paste" method for stretchable 3D antenna

Buckling instabilities have been used to process traditional inorganic materials to gain structural stretchability. For example, out-of-plane structures have been achieved by patterning and transfer printing planar films onto a pre-strained substrate followed by releasing the strain [64–69]. Films patterned by the "cut-and-paste" method can be combined with the buckling design to enhance the stretchability and even optimize functional performances. For example, Yan et al. [28] reported a stretchable and shape-controllable 3D antenna fabricated by the "cut-transfer-release" method. As illustrated in Figure 2(a), a polyimide thin film was mechanically carved with the pattern of an antenna followed by Au deposition. After removing excess region, the patterned antenna can be transferred on a pre-strained elastomeric substrate. After releasing the pre-strain, the planar antenna would buckle into a 3D antenna. Upon stretching, the 3D antenna can recover to a planar antenna without mechanical failure, as shown in Figure 2(b). In this work, the frequency of this particular antenna design was not very sensitive to the mechanical deformation. However, this work points to a route of building 3D reconfigurable antenna.

2.3 "Cut-and-paste" method to pattern ceramic materials

Although metal and polymer sheets are cuttable due to their plastic deformability, radial micro-cracks would form as soon as a mechanical blade lands on a ceramic film, similar to the micro-indentation induced cracks in brittle films [70]. Therefore, a slightly modified "cut-deposit-paste" method has been developed for patterning ceramic thin films using mechanical cutter plotters [54]. Indium tin oxide (ITO) was used as a representative ceramic film because of its electrical conductivity, which can reflect its mechanical integrity. The "cut-deposit-paste" process is illustrated by Figure 3(a)–(i). A piece of 13-μm-thick transparent PET film was attached to a TRT, which was supported by a cutting mat (Figure 3(a)). Without ITO coating yet, the PET can be carved with the designed serpentine patterns (Figure 3(b)). After removing the TRT-supported PET from the cutting mat (Figure 3(c)), 200-nm-thick ITO was sputtered on PET at room temperature (Figure 3(d)). Although the target substrate Tegaderm tape has adhesive, depositing 5-nm-thick Ti and 50-nm-thick

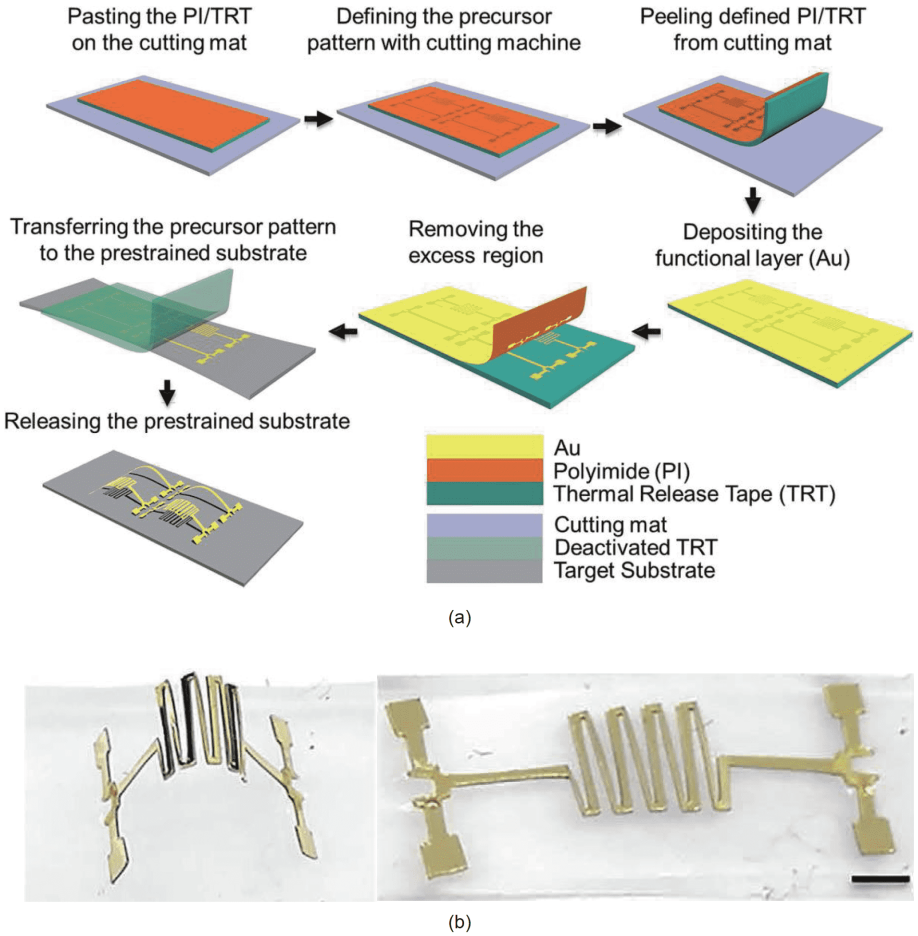

Figure 2 (Color online) (a) The fabrication process of the "cut-transfer-release" method; (b) the tunable 3D antenna made out of the "cut-transfer-release"

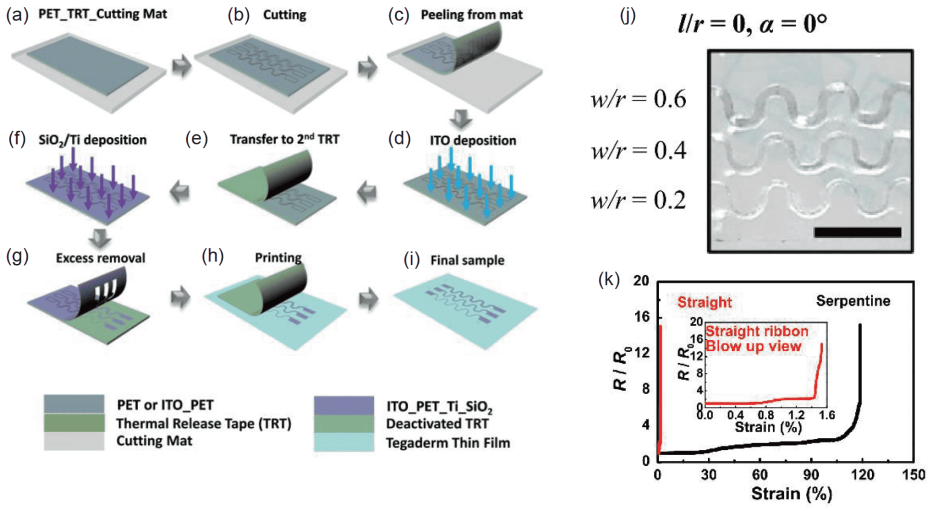

Figure 3 (Color online) Schematics of the "cut-deposit-paste" process of fabricating stretchable ITO serpentines on stretchable Tegaderm substrates. (a) Laminating a 13-μm-thick PET on a TRT on a cutting mat; (b) using a mechanical cutter plotter to carve out serpentine-shaped seams on PET; (c) peeling TRT-supported PET from the cutting mat; (d) sputtering 200-nm-thick ITO on PET; (e) transferring ITO covered PET from one TRT to another with the backside of PET exposed; (f) sputtering 5-nm-thick Ti and 50-nm-thick SiO_2 on the backside of the PET (for weakly-bonded serpentines, this step is skipped); (g) removing unwanted PET, leaving only ITO_PET serpentines on the TRT; (h) pasting the ITO/PET serpentine ribbons on Tegaderm with ITO facing up; (i) the final sample; (j) a picture of a weakly-bonded ITO/PET displays good transparency, the scale bar is 5 mm; (k) electrical resistance as a function of the applied uniaxial tensile strain for straight and serpentine ITO ribbons. Republished with permission from Extreme Mechanics Letters [54].

SiO_2 on the other side of the PET can further enhance the adhesion (Figure 3(e)–(f)). Finally, after removing the excess regions (Figure 3(g)), the Ti/SiO_2-coated PET side was pasted on the target Tegaderm tape (Figure 3(h)–(i)). A picture of the as-fabricated ITO/PET serpentine ribbons without Ti/SiO_2 adhesion promoter layer is displayed in Figure 3(j) where the transparency of the ITO is obvious. No micro-cracks can be observed at the edge of the ITO because ITO was deposited after the PET was cut. Peeling off the excesses did not cause any damage to the leftover ITO because the as-deposited ITO was already discontinuous at the seams. *In situ* electrical resistance measurement during uniaxial tension tests (Figure 3(k)) indicates the strain-to-rupture can increase from 1% to 120% after patterning ITO into serpentine shapes. This is the first time that brittle ITO can be stretched beyond 100%. Such "cut-deposit-paste" method is expected to be also applicable to other brittle films as long as they can be deposited at a temperature acceptable to the polymer support.

3 "Cut-and-paste" process for atomically thin materials and their assemblies

Except for ITO, graphene, a single layer of carbon atoms arranged in a honeycomb lattice, is also a transparent conductor [71]. In fact, graphene belongs to an emerging family of atomically thin materials called 2D materials, which can be used as conductors, semiconductors and dielectrics in future atomically thin electronics [72]. Some of their capabilities such as electrical and thermal conductivity can further be enhanced by layer-by-layer assembly of 2D materials [73–79]. Notably, 2D materials with atomic thinness, such as graphene, monolayer or few-layer MoS_2 and many more, are intrinsically more deformable than their bulk counterparts and other conventional electronic materials [80–88]. As a result, 2D materials do not suffer from brittle fracture like the ceramic thin films when subjected to mechanical identation or cutting [89]. Besides, 2D materials have many other merits such as optical transparency [90], electrochemical inertness [91], and biocompatibility [92], which make them ideal building blocks for flexible electronics [72] and bio-integrated electronics [93,94]. Although 2D materials can be patterned by photo- or e-beam lithography, every chemical in the process would inevitably leave residuals on the surfaces of the 2D materials, which have a dominating effect on their properties and performance [95]. As a result, a dry patterning process like the "cut-and-paste" method is highly desirable for fabricating devices based on 2D materials. In this section, we discuss the application of "cut-and-paste" method in fabricating graphene-based devices. We also expect that the "cut-and-paste" method can be applied to other 2D materials or their stacks in the future.

3.1 Graphene e-tattoos (GETs)

Our group built the first sub-micron-thick transparent and imperceptible graphene e-tattoos (GETs) through a "wet transfer, dry patterning" process where the dry patterning was done through the "cut-and-paste" method [55]. The

process is illustrated in Figure 4(a). Following well-established procedures [96], we used chemical vapor deposition (CVD) to grow large area monolayer graphene on Cu foils. We applied a popular "wet transfer" method to separate monolayer graphene from the Cu foil [97]. First, we protected monolayer graphene with spin-coated sub-micron-thick poly(methyl methacrylate) (PMMA). Then we chemically etched the Cu without causing mechanical damage to the graphene. Our process differs from conventional wet transfer process after Cu etching. Conventionally, the PMMA-protected graphene is scooped by a target substrate with PMMA facing up. After dissolving PMMA by acetone, monolayer graphene is left on the target substrate for subsequent fabrication or characterization. In our process, PMMA was not removed because it can serve as a permanent mechanical support for graphene even after the tattoo was transferred on human skin. As a result, we used a commercial tattoo paper which comes with water-soluble adhesive to fish graphene/PMMA with graphene facing up. We then used the mechanical cutter plotter to carve out the designed filamentary serpentine mesh with 0.9-mm ribbon width on the graphene/PMMA bilayer. After manually removing the excess materials, the GET can be directly pasted on human skin exactly like a temporary transfer tattoo, with graphene in direct contact with the skin. The total thickness of the GET was dominated by the PMMA layer, which was measured to be only 463 ± 30 nm. This thickness gives the GET an ultra-low bending stiffness of 2.7×10^{-11} N m, which is one order smaller than that of an Au-based e-tattoo (e.g., 3.49×10^{-10} N m for 100-nm-thick Au on 700-nm-thick polyimide). As a result, without using any adhesives, the GET was able to fully conform to the microscopically rough skin surface even under severe skin deformation, as evidenced in Figure 4(b). Meanwhile, the optical transmittance of the GET was 84% to 88% within the visible spectrum. We have demonstrated that the GET can fully replace the Au electrodes for the measurement of ECG, EEG, EMG, and skin hydration. The GET-skin interface impedance and signal-to-noise ratio (SNR) were found to be on par with wet gel electrodes, which have limitations including skin irritation and wearable time.

We also created an imperceptible human-robot interface (HRI) by applying GETs as the invisible and imperceptible

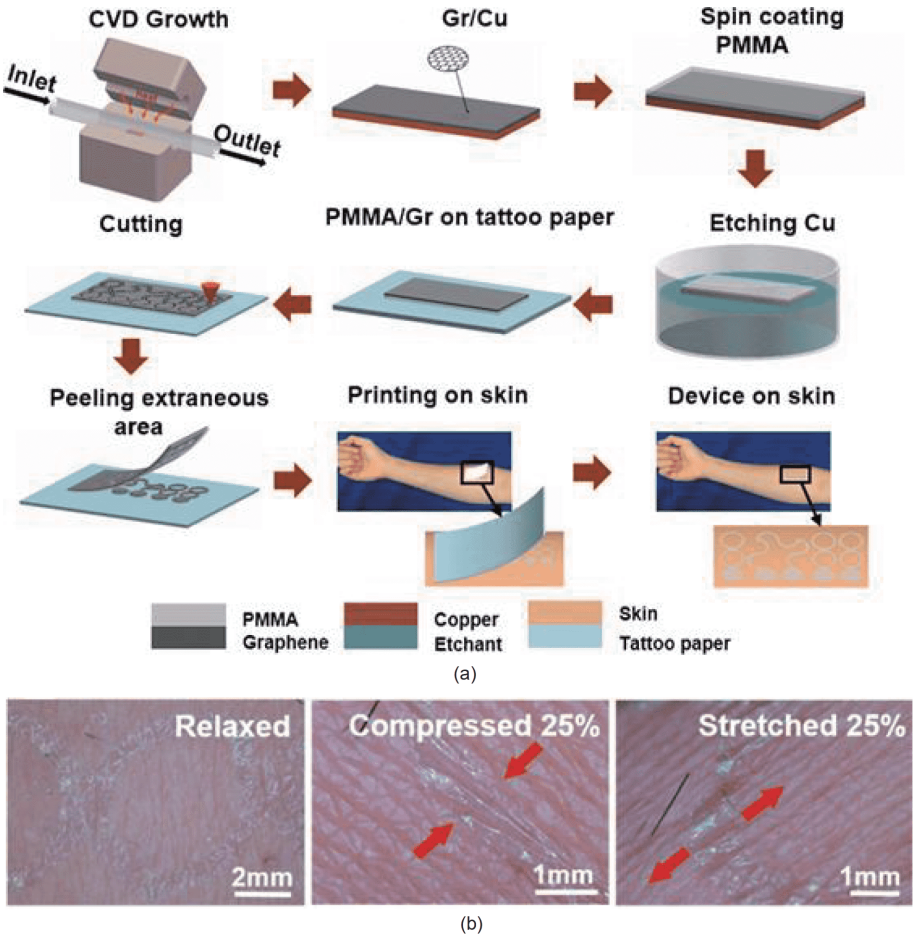

Figure 4 (Color online) (a) "Wet transfer, dry patterning" fabrication of GETs, which involves the "cut-and-paste" process. Instead of pasting on a polymer substrate, GETs can be directly pasted on the skin without adhesives due to their sub-micron thickness. (b) Micrographs of a transparent GET on relaxed,

surface electrodes on human face, specifically around human eyes. By wirelessly measuring and interpreting the electro-oculogram (EOG), we achieved real-time control of a drone by eyeball rotation [20].

3.2 Graphene antennas

In the previous section, we have proved that large-area monolayer graphene can be used as effective electro-physiological electrodes for high fidelity electro-physiological sensing thanks to their low electrode-skin interface impedance. However, it is well known that the sheet resistance of monolayer graphene is too high [98] to serve as effective interconnects or antenna coils. Alternatively, bottom-up graphene assemblies such as "graphene paper" (Figure 5(a)) could be made into good conductors with low cost, high mechanical reliability, and good chemical stability [77,99,100]. Graphene paper is also mechanically cuttable. For example, Scidà et al. [56] fabricated flexible near field communication (NFC) antenna by essentially the "cut-and-paste" method (Figure 5(b)). Benefiting from the mechanical robustness of graphene and its assemblies, the flexible antenna showed trivial changes in the electro-magnetic characteristics after 1 million cycles of bending. Compared with a metal-based commercial antenna, the graphene-paper-based antenna is more bendable and cyclable, which can be attributed to the limited plasticity in the graphene paper. Expectedly, this combination of bottom-up assembly and top-down cut-and-paste process is readily extendable for the fabrication of soft electronics based on the emerging 2D materials, or more broadly, nanomaterials including nanotubes and nanowires [101–105].

4 Cut-solder-paste method

To improve the practical use of soft electronics such as the e-tattoos, extensive efforts go into extending them with active functionalities such as amplification and wireless communication, which involves IC integration on the device. The "cut-and-paste" method can offer adequate spatial resolutions for thin film devices to be compatible with commercially available ICs. This section discusses our recent achievement in the rapid prototyping of NFC-enabled battery-free wireless e-tattoos [106] using a "cut-solder-paste" method [52]. The process is illustrated in Figure 6(a)–(f). An 18-μm-thick Cu foil was used to fabricate the stretchable NFC antenna and the interconnects. As many soft substrates including Tegaderm tapes are not able to survive soldering temperature, IC soldering has to be finished before the pasting step. To enable soldering, the cut-patterned Cu circuit was transferred onto a water-soluble tape (WST) backed by a Kapton tape. Soldering off-the-shelf NFC ICs and other discrete components can be readily completed on such a bilayer backing system. After soldering, the circuit can be successfully released and pasted onto the target substrate by dissolving the WST using water. The cutting process could be done within several minutes, and the subsequent soldering, transferring and laminating process would take about 20 to 60 min, depending on how complicated the circuit is. Figure 6(g) displays a picture of the wireless e-tattoo on skin being twisted. A light emitting diode (LED) on the e-tattoo was wirelessly turned on by an interrogator placed 3 cm above the e-tattoo. We have also demonstrated that thermistors and photodiodes integrated on the e-tattoo can wirelessly transmit data to an NFC-enabled smart-phone.

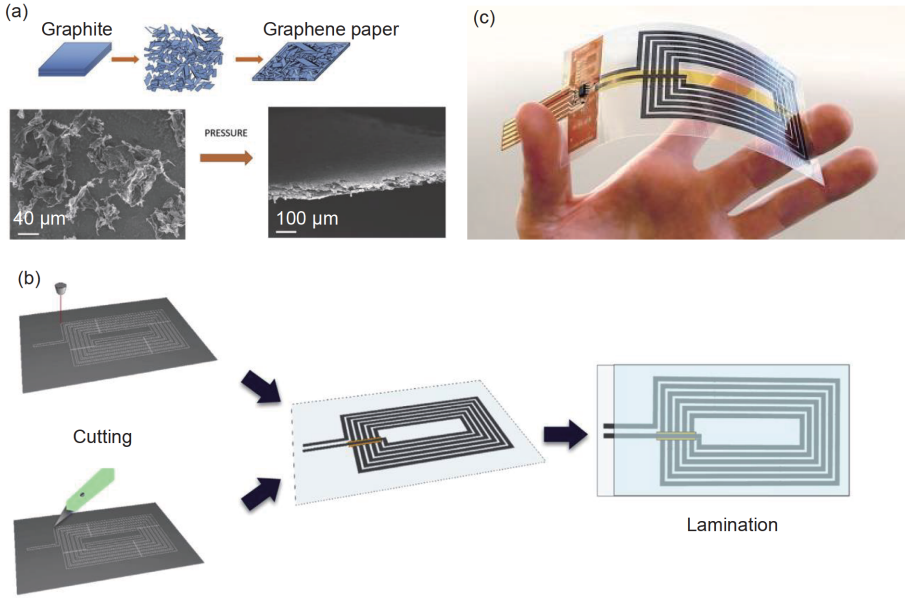

Figure 5 (Color online) (a) Graphene paper fabrication process and scanning electron microscope (SEM) pictures; (b) fabrication process of the graphene-paper-based NFC antenna; (c) a picture of the antenna under bending. Republished with permission from Materials Today [56].

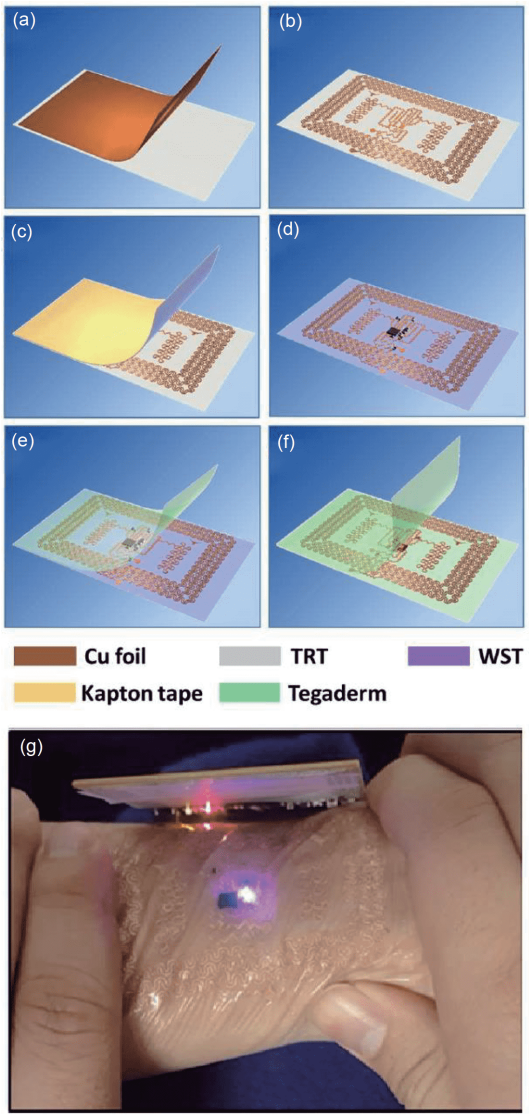

Figure 6 (Color online) Schematic illustrations of the "cut-solder-paste" process. (a) Cu foil laminated on TRT; (b) cut-patterned Cu antenna and interconnects; (c) transfer of Cu circuit onto a water-soluble tape (WST) backed by a Kapton tape; (d) solder NFC chip, thermistor, LED, photo-transistor, and other discrete components on the Cu circuit; (e) dissolving WST to paste the completed circuit to Tegaderm; (f) sandwich the circuit with another Tegaderm; (g) an NFC-enabled e-tattoo twisted on skin while still being wirelessly turned on. Republished with permission from 2017 39th Annual International Conference of the IEEE Engineering in Medicine and Biology Society (EMBC) [106].

These results demonstrated the capability of the "cut-solder-paste" method in integrating rigid ICs on the stretchable platform. With such development, the cut-and-pasted e-tattoos are one step closer to real-life applications.

5 Conclusions

Soft electronics including flexible and stretchable electronics have demonstrated many unprecedented capabilities expanding into healthcare, communication, soft robotics, and consumer markets. It thus creates an unignorable demand for improved manufacturability in terms of cost, yield, throughput, and versatility. Instead of using conventional wafer-based microelectronics fabrication approach, the "cut-and-paste" technologies summarized in this review offer a more cost- and time-effective solution for the rapid prototyping of soft electronics of various materials and functionalities. It complements the additive manufacturing in the digital manufacture space even for scaled-up production. Due to the simplicity in facility and condition, it is also friendly to be added to other manufacturing processes for building more diverse and exciting hybrid soft electronics in the future.

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